

Application No. 10/535,050  
Harbec et al.

Docket number: 1770-322US

### AMENDMENTS TO THE SPECIFICATION

Please replace paragraph [0005] as amended with the response filed on November 12, 2009 with the amended paragraph, as follows:

[0005] This disclosure relates to a process for the manufacture of carbon nanostructures, from carbon nanotubes and carbon non-onions, comprising the steps of a) providing a high enthalpy metal electrode generated direct current thermal plasma torch having a plasma forming gas feed and a cooled nozzle attached thereto, the torch being connected to a cooled reactor having a quenching zone downstream of the plasma torch for the formation of carbon nanostructures; b) selecting a catalyst metal and providing the catalyst metal to the plasma stream, selecting a torch power at a level of from about 30 kW up to a multi-megawatt level, selecting a flow rate for the plasma forming gas feed, and selecting the reactor pressure so as to provide a plasma torch stream temperature required to vaporize and maintain the selected catalyst metal in the vapor state; c) providing a feed of a carbon containing substance and a carrier gas at a selected flow rate to the plasma stream; and d) the resulting plasma stream containing carbon, carrier gas and metal vapor entering the quenching zone of carbon nanostructure formation, wherein the plasma stream is rapidly cooled at a quenching rate which can be calculated in accordance with the formula  $\Delta T/t$ , where  $\Delta T$  is the temperature difference between the temperature of the plasma entering the nozzle  $T_2$  and the temperature of the plasma in the quenching zone  $T_1$ , with the average

temperature entering the nozzle  $T_2$  being calculated by the formula  $T_2 = T_1 + \frac{W_p}{\dot{m}C_p}$

where  $T_1$  is room temperature;  $W_p$  is the energy input to the plasma,  $\dot{m}$  is the mass flow rate of the carrier gas;  $C_p$  is the specific heat of the carrier gas; and  $t$  is the time for the plasma stream to travel from the plasma torch to the quenching zone, where  $t$  can be calculated by the formula  $t = \text{length of nozzle} / \text{velocity of plasma gas entering the nozzle}$ , whereupon metal catalyst nanoparticles acting as nucleation sites and catalyst for the growth of carbon nanostructures are generated *in situ* in a diameter range of from about 2 to about 30 nm from the metal catalyst vapor, which with atomic carbon from the carbon containing substance, form such structures in a diameter range of from about 2

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to about 30 nm, which carbon nanostructures are then collected. In a further aspect, the process involves the manufacture of carbon nanostructures, the carbon nanostructures being selected from carbon nanotubes and carbon nano-onions, comprising the steps of a) providing a high enthalpy metal electrode generated direct current thermal plasma torch having a plasma forming gas feed and a cooled nozzle attached thereto, the cooled nozzle having a carbon containing substance and carrier gas feed, the torch being connected to a cooled reactor having a quenching zone downstream of the plasma torch for the formation of carbon nanostructures; b) selecting a catalyst metal, selecting the torch power at a level of from about 30 kW up to a multi-megawatt level, selecting the flow rates of the plasma forming gas feed and the carbon containing substance and carrier gas feed, and selecting the reactor pressure so as to provide a plasma torch temperature required to vaporize the catalyst metal and maintain the catalyst metal in vapor form, a plasma stream expansion at the nozzle exit and the downstream quenching zone allowing cooling of the plasma stream wherein the plasma stream is rapidly cooled at a quenching rate which can be calculated in accordance with the formula  $\Delta T / t$ , where  $\Delta T$  is the temperature difference between the temperature of the plasma entering the nozzle  $T_2$  and the temperature of the plasma in the quenching zone  $T_1$ , with the average temperature entering the nozzle  $T_2$  being calculated by the formula  $T_2 = T_1 + \frac{W_p}{\dot{m}C_p}$ , where  $T_1$  is room temperature;  $W_p$  is the energy input to the plasma,  $\dot{m}$  is the mass flow rate of the carrier gas;  $C_p$  is the specific heat of the carrier gas; and  $t$  is the time for the plasma stream to travel from the plasma torch to the quenching zone, where  $t$  can be calculated by the formula  $t = \text{length of nozzle} / \text{velocity of plasma gas entering the nozzle}$ , to generate *in situ* nanometer sized metal catalyst particles, which act as catalyst and nucleation sites for the formation of carbon nanostructures; and c) injecting the carbon-containing substance and carrier gas into the nozzle at a feed rate that allows the formation of atomic carbon, and injecting the resulting plasma stream seeded with atomic carbon and metal vapours into the quenching zone downstream of the plasma torch which, in the presence of the nanometer sized metal catalyst particles generated *in situ* having a diameter of from about 2 to about 30nm, form carbon nanostructures having a diameter of from about 2 to about 30nm, which are then collected. In another aspect, the process for the

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manufacture of carbon nanostructures, the carbon nanostructures being selected from carbon nanotubes and carbon nano-onions, comprising the steps of a) selecting tungsten as a catalyst metal and providing a high enthalpy tungsten-coated electrode in a direct current thermal plasma torch having an inlet for a plasma forming gas feed at a flow rate of about 100 to about 225 standard litres per minute; b) selecting the torch power at a level of from about 30 to about 65kW and the reactor pressure at about 200 to about 800 torr, so as to provide a plasma torch temperature required to vaporize the tungsten-coated metal electrode and maintain the tungsten metal in the form of a vapor; c) selecting a tungsten nozzle attached to the torch outlet and cooled to a temperature below 1500°C, the nozzle having a carbon containing substance and a carrier gas feed inlet and injecting the carbon-containing substance at a rate of about 0.15 mol/min with a carrier gas at a flow rate of about 20 standard litres per minute into the plasma stream from the nozzle inlet; and d) using the cooling of the plasma stream above  $10^7^\circ\text{C/s}$  produced by the carbon-containing substance and carrier gas feed, and by a supersonic shock created at an exit of the nozzle or the provision of an expansion in the nozzle internal diameter, wherein the plasma stream is rapidly cooled at a quenching rate which can be calculated in accordance with the formula  $\Delta T/t$ , where  $\Delta T$  is the temperature difference between the temperature of the plasma entering the nozzle  $T_2$  and the temperature of the plasma in the quenching zone  $T_1$ , with the average temperature

entering the nozzle  $T_2$  being calculated by the formula  $T_2 = T_1 + \frac{W_p}{mC_p} t$ , where  $T_1$  is

room temperature;  $W_p$  is the energy input to the plasma,  $m$  is the mass flow rate of the carrier gas;  $C_p$  is the specific heat of the carrier gas; and  $t$  is the time for the plasma stream to travel from the plasma torch to the quenching zone, where  $t$  can be calculated by the formula  $t = \text{length of nozzle} / \text{velocity of plasma gas entering the nozzle}$ , to generate *in situ* nanometer sized tungsten catalyst particles having a diameter of from about 2 to about 30nm, which act as the catalyst and nucleation sites for the formation of carbon nanostructures having a diameter of from about 2 to about 30nm within the plasma stream, which are then collected.